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A year–long comparison of particle formation events at paired urban and rural locations

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ABSTRACT

Ultrafine particle size distribution data were collected in downtown Toronto and rural Egbert from May 2007 to May 2008. Particle formation events were observed in both locations and contributed to increased concentrations of particles less than 25 nm in diameter. These events were more frequent in spring and fall and rarely occurred in winter. Stronger solar radiation and drier air were correlated with the occurrence of formation events at both locations. Nucleation events occurred simultaneously at both sites on 10% of the days, and these events involved a shared air mass. Half of these simultaneous events were associated with northern air masses and only a quarter with southerly air masses. The higher loading of aged particles in southerly air masses transported from upwind industrial sectors appeared to limit the occurrence of nucleation events. Formation events occurred less frequently in downtown Toronto than at the rural site, and the frequency was lower on weekdays. It is hypothesized that vehicular emissions were responsible for the suppression of nucleation events in downtown Toronto.

Keywords: *Particle formation, ultrafine particles, traffic emissions, particle nucleation and growth, condensation sink*

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1. Introduction

Atmospheric particles exert considerable climate influence and affect human health. In the global troposphere, aerosol particles influence the Earth's radiation budget by directly absorbing or scattering solar radiation and indirectly acting as cloud condensation and ice nuclei. Particles also serve as an interface where heterogeneous reactions occur (Seinfeld and Pandis, 2006). Further these particles can reduce visibility and adversely impact human health. Epidemiological studies have linked cardiovascular or respiratory morbidity and mortality to particulate matter exposure (Peters et al., 1997; Oberdorster et al., 2002). In order to estimate and predict the environmental and health impacts of aerosol particles, it is essential to understand their genesis and evolution in the atmosphere. One of the key processes in this regard is the formation of ultrafine particles and their subsequent growth through nucleation events. These events can produce sharp increases in the number concentration of particles and have been observed in a range of environments throughout the globe. Specifically, new particle formation events have been seen in regions spanning sub–Arctic Lapland (Vehkamaki et al., 2004), boreal forest in Finland (Kulmala et al., 1998; Dal Maso et al., 2005), and urban areas in Europe (Alam et al., 2003; Hamed et al., 2007; Salma et al., 2011), North America (Jeong et al., 2004; Stanier et al., 2004; Jeong et al., 2006; Qian et al., 2007), and Asia (Wehner et al., 2004; Monkkonen et al., 2005). Nevertheless, the mechanisms underlying these events have not been fully elucidated, mainly due to the challenges in directly measuring particles with diameter around 1.0 nm and analyzing the chemical composition of these newly formed particles.

Insight into these mechanisms has been gained through laboratory experiments and field studies. Sulfuric acid is considered a key nucleation precursor due to its low equilibrium vapor pressure in the atmosphere (Seinfeld and Pandis, 2006). Laboratory–based studies have often focused on connecting the concentration of precursor gases, such as sulfuric acid, to nucleation rate (Sipila et al., 2010). Moreover, the role of ammonia and organics have been studied through a number of experiments since binary nucleation of sulfuric acid and water failed to reproduce the nucleation rate measured in the field (Metzger, et al., 2010; Benson et al., 2011). In field studies, researchers have examined the atmospheric conditions favoring nucleation events. Boy and Kulmala (2002) analyzed the influences of meteorological parameters and reported that new particle formation was correlated with solar radiation and anti–correlated with relative humidity (RH). The observation of nucleation events was also found to be negatively correlated with high concentrations of pre–existing particles in field measurements (Weber et al., 1997).

Field studies conducted at multiple locations, either simultaneously or consecutively, have examined the influence of local meteorology, air masses, geography, and anthropogenic emissions. Vana et al. (2004) found that nucleation events at multiple locations in northern Europe were associated with cold Arctic air masses. An extensive field measurement campaign was conducted at 12 locations with varying environments in Europe, and nucleation at different sites was found to follow different seasonal patterns (Manninen et al., 2010). Paasonen et al. (2010) further reported that a correlation between the nucleation rate and the concentration of precursor gases varied between locations,

suggesting that different chemical compounds might contribute to nucleation at different locations.

Jeong et al. (2010) analyzed three weeks of particle number (PN) concentrations and size distributions obtained at five urban and rural locations in southern Ontario, Canada. These authors found that both anthropogenic and biogenic sources contributed to nucleation and growth of particles at locations situated close to industrial districts, and that these sources play an important role in determining aerosol population at both rural and urban sites. These findings pointed to the need for a longer–term study, to further resolve the extent to which differing emissions contribute to particle formation and growth at urban and rural locations. In this follow–up study, particle size distribution measurements were collected simultaneously for one year (May 2007–2008) in downtown Toronto and rural Egbert. These data were used to compare particle nucleation and growth events at these sites. It was hypothesized that examining nearby sites with differing mixes of anthropogenic and biogenic emissions would help elucidate the effects of these emissions, and shared parameters such as meteorology and air mass origin, on the occurrence of particle formation.

2. Experimental Methods

2.1. Monitoring sites and data sources

Toronto. Ultrafine particle number concentrations were measured in ambient air sampled at the laboratory of the Southern Ontario Centre for Atmospheric Aerosol Research (SOCAAR). SOCAAR is located at the Wallberg Building at the University of Toronto in downtown Toronto, Ontario, Canada (43.66° N, 79.40° W), and surrounded by multi–story buildings (Figure 1). The inlet is 15 m away from College Street, which experiences a traffic volume of ~20 000 vehicles per day. Particle size distributions were measured by a Scanning Mobility Particle Sizer (SMPS, TSI, St. Paul, MN) equipped with a nano– Differential Mobility Analyzer (DMA, TSI 3085, St. Paul, MN) and Ultrafine Water–based Condensation Particle Counter (UWCPC, TSI 3786, St. Paul, MN). The SMPS detected particles with mobility diameters of 3 to 106 nm every 2 minute. In addition, a TSI Fast Mobility Particle Sizer (FMPS) was employed to obtain particles with mobility diameters of 6 to 560 nm every second (Table 1). The FMPS data were used when the SMPS data were not available for Toronto. The FMPS data were corrected due to multiple charging of particles from 8 to 100 nm (Jeong and Evans, 2009). Also, the size distributions of particles larger than 100 nm were corrected based on polystyrene latex (PSL) calibration particles and a comparison with the SMPS equipped with a long DMA (TSI 3081, St. Paul, MN) used for Toronto. The SMPS and FMPS data were well–correlated after the FMPS data were corrected (Jeong and Evans, 2009).

A SmartEye Traffic Data Sensor (TDS) was employed to measure traffic volume along College St. (four–lane street). The TDS was set up on the roof of the Gage building situated 150 m west of the SOCAAR site. The TDS uses edge–detection to count the number of vehicles across all four lanes. The TDS continuously recorded traffic volume every five minute. The traffic data used here were collected between June and December 2010, several years after the ultrafine particle data was collected. Further the data from this traffic sensor had known limitations, including undercounting at night. However, despite these limitations, this traffic data was sufficiently representative of diurnal and seasonal traffic patterns in downtown Toronto to meet the needs of this study (Sabaliauskas et al., 2012).

Trace gas concentrations were obtained from the Ontario Ministry of the Environment (MOE) downtown site, situated approximately 850 m northeast of the SOCAAR sampling site (MOE, 2012). This MOE site provided hourly averaged concentrations of sulfur dioxide (SO_2), nitric oxide (NO), nitrogen dioxide (NO_2), carbon monoxide (CO), ozone (O_3), and mass concentrations of fine particulate matter ($PM_{2.5}$). The meteorological data were obtained from Environment Canada (EC) at the Pearson International Airport located approximately 20 km west of the SOCAAR site. This EC site provided hourly temperature (T), relative humidity (RH), wind speed (WS), and wind direction (WD). Solar radiation data were taken from the University of Toronto Mississauga Campus' meteorological station. Though this site is situated 25 km west of the SOCAAR site, its solar radiation data were the most consistently available throughout the campaign.

Egbert. The rural data were collected near Egbert Ontario, at Environment Canada's Centre for Atmospheric Research and Experiment (CARE). CARE is located approximately 80 km north of the Toronto site (44.23° N, 79.78° W) and is surrounded by mixed forest and farmland (Figure 1). The nearest road to the sampling, located 75 m away, experiences only a few vehicles per hour. Particle size distributions between 10 and 400 nm were detected every 15 minute by a SMPS equipped with a long DMA (TSI 3081, St. Paul, MN) and CPC (TSI 3025, St. Paul, MN). The centre also provided meteorological and traces gas concentration data except $PM_{2.5}$ data; $PM_{2.5}$ data were obtained from Barrie, a nearby city 15 km northeast of Egbert. While the sampling location in Toronto was heavily influenced by anthropogenic emissions, such as vehicle exhaust, Egbert experienced minimal local emissions. However, both locations were at times impacted by air masses from the south and southwest, containing outflow from industrialized regions in southwestern Ontario and mid–western United States.

Figure 1. *Map of Toronto and Egbert. Egbert is located 80 km north of Toronto and experiences minimal anthropogenic emissions.*